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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In the Application of:

Andrea F. Gulla, et al.

Serial No.: 10/830,182

Filed: April 22, 2004

For: CATALYST ... REDUCTION

: Group: 1755

: Examiner: Hailey, Patricia L.

Hedman and Costigan  
1185 Avenue of the Americas  
New York, NY 10036  
May 22, 2008

BRIEF ON APPEAL

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

REAL PARTY IN INTEREST

The real party in interest in the above application is Industrie De Nora S.p.A. of  
Milan, Italy.

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**RELATED APPEALS AND INTERFERENCES**

There are no other prior and pending appeals, interferences or judiciary proceedings which may be related to, directly affect or be directly affected by or have a bearing on the Board's decision.

**STATUS OF THE CLAIMS**

Claims 41 to 45 stand allowed and Claims 1, 3, 5 to 7, 10, 12 to 22, 36 to 40 and 46 to 48 stand rejected and are on appeal, all other claims being cancelled.

**STATUS OF THE AMENDMENTS**

No amendment was filed subsequent to the final rejection of December 20, 2007 although a pre-appeal brief request for review was filed on March 24, 2008 which was decided on April 24, 2008 to proceed to the Board of Appeals and Interferences without change.

SUMMARY OF THE CLAIMED SUBJECT MATTER

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Claim 41 is drawn to a method for producing a gas diffusion electrode of a gas diffusion electrode of a conductive carbon black web with an electrocatalyst comprised of a cobalt-ruthenium sulfide supported on carbon wherein the conductive web is coated on at least one side with the catalyst, subjecting the coated web to a sintering step and then heating under an inert atmosphere (page 9, lines 3 to 20 and lines 1 to 6 of page 10).

Claim 45 is directed to hydrochloric acid depolarized electrolysis process using an electrode produced by the process of class 41 as the cathode in an electrolysis cell fed with a hydrochloric acid aqueous solution and oxygen while applying an electric current (page 14 last 3 lines and lines 1 to 17 of page 15).

These claims were allowed on the basis that "the cited references do not teach or suggest the limitations of claims 41 to 46 (page 9 of June 15, 2006 office action). In the office action of June 28, 2007, the Examiner stated the "prior art of record does not teach or suggest the step of sintering in a hydrogen atmosphere, followed by subsequent heating under inert atmosphere".

Claim 1 is directed on electrocatalyst for oxygen reduction comprising a cobalt-ruthenium sulfide supported on a conductive carbon black (line 16 of page 6 to line 2 of page 7 and claim 20 is directed to a gas diffusion electrode comprising a conductive web with a catalyst of claim 1 applied or at least one face of said conductive web lines 11 to

14 of page 6 and lines 2 to 10 of page 9). The advantages of the invention are shown in Examples 4 to 6.

### GROUNDS OF REJECTION

Claims 1, 3, 5 to 7, 10, 12, 13, 15 to 29 and 46 to 48 were rejected under 35 USC 103 as being obvious over the Forquy patent taken in view of the Lang et al and Ito et al patents and claims 1, 3, 5 to 7, 12, 14 to 19, 47 and 48 were rejected under 35 USC 103 as being obvious over the Kobylinski et al patent taken in view of the Forquy et al patent. The Examiner states that Forquy discloses a catalyst comprised of ruthenium sulfide and at least one other transition metal including cobalt on a support. The Examiner concedes Forquy does not disclose a "conductive carbon black support" but cites Lang et al as showing a catalyst on a "preformed carbon support" including carbon black and the Examiner concludes that carbon black and active carbon are known catalyst supports. Kobylinski et al is cited as showing a catalyst by coating a support with a ruthenium or rhodium sulfide with a platinum metal sulfide. The Examiner concedes it does not teach electrocatalyst or a conducive support but it would be obvious to have this. The Examiner concedes cobalt sulfide is not taught but cites Forquy as showing this.

APPLICANTS' ARGUMENTS

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Applicant respectfully requests the Board of Patent Appeals and Interferences to reverse the Examiner's rejections since the Forquy et al patent, taken alone or in combination with the Lang et al and Ito or Kobylinski et al patent, does not anticipate or render obvious Applicants' invention. As new as Applicants can determine, neither the Examiner nor the Appeal conference board have responded to Applicants' arguments that the Professor Faita's declaration clearly shows that Forquy et al is unrelated to Applicants' invention.

The Forquy patent rejection is based on the Examiner's assumption that an active carbon is per se a conductive support and therefor is inherently suitable for use as an electrocatalyst support which is obviously wrong. Secondly, Forquy et al. simply does not anticipate our invention, which is directed to a carbon black supported cobalt and ruthenium sulfide. Forquy et al is directed to ruthenium sulfide on any possible inert support, optionally comprising at least one other transition metal selected from a long list. As far as novelty is concerned, claim 1 is at the very least to be regarded as a purposive selection over Forquy et al, which therefore does not anticipate the invention.

As regards to inventiveness, Forquy et al belongs to an unrelated field of application (heterogeneous catalysis vs. electrocatalysis, and dehydrogenation vs. oxygen reduction) that one skilled in the art would never have contemplated selecting such document to derive any useful teaching for making an electrocatalyst for oxygen

reduction. All grounds of rejection based on Forquy et al are hence moot.

The Lang et al reference, even if combined with Forquy patent is again directed to a catalyst having hydrogenation activity and would not be considered by one skilled in the art wishing to solve the technical problem of providing an electrocatalyst for oxygen reduction resistant to a chlorinated hydrochloric environment. Neither electrocatalysis or oxygen reduction are mentioned in Lang et al., nor the specific embodiment of ruthenium sulfide is the object of a specific example.

But the sentence which clearly demonstrates how far is the Examiner from appreciating the technical content of the invention is to be found on page 3 of the Office Action, referring back to Forquy's patent:

"The catalyst of Forquy's et al. would be expected by one of ordinary skill in the art to function as an "electrocatalyst for oxygen reduction", absent the showing of convincing evidence on the contrary (!) first of all Prof. Faita's declaration is evidence of the contrary, and secondly one considering the non conductive catalyst for THT dehydrogenation of Forquy et al. as a viable candidate to function in oxygen reduction electrocatalysis, would per se show to lack any skill whatsoever in the art of electrocatalysis, given the fact that electrical

conductivity is one of the key points to be taken into consideration.

To summarize:

- \* the invention belongs to the field of electrocatalysis, in particular to electrocatalysts for oxygen reduction.
- the closest prior art consists of rhodium sulfide supported on conductive carbon black, which is active and stable in such an electrolytic environment but that is way too expensive.
- the solution provided by the present invention consists of a ruthenium-cobalt sulfide supported on a conductive carbon black, as claimed in present claim 1;

After utility and novelty are established, section 103 makes patentability depend upon non-obviousness of the invention to be patented. The first sentence of this section requires that:

- (1) the scope and content of the prior art be determined;
- (2) the differences between the prior art and the claims in issue be ascertained;
- (3) the level of ordinary skill in the pertinent art be resolved

Referring to item (1), scope and content of the pertinent

art of electrocatalysis must be determined, that is of a science dealing with developing electrically conductive catalysts to enhance electrochemical reactions in suitable cells and electrolyzers. One of skill in the art would therefore have taken into consideration US Patents No. 5,958,197 and No. 6,149,782 (cited in the application), which disclose electrocatalysts for oxygen reduction in a hydrochloric environment. Additionally, of all the documents cited by the Examiner, one of skill in the art would only have considered the Reeve et al. article, which is directed to electrocatalysts. At best, he would also have considered searching technical disclosures dealing with stability of noble metal compounds in a chlorinated hydrochloric environment.

The Examiner has conceded finally that Forquy does not explicitly disclose carbon black as the support nor a surface area for the support. The important thing is Forquy does not teach any support for the catalyst which is electrically conductive as required by the claims and is no way directed to Applicants' problem and Applicants' solution thereof. A non-conductive support could not be deemed equivalent to Applicants' electrically conductive carbon black.

With respect to Claims 20 to 22 and 36 to 40 which have been rejected under 35 USC 103 as being obvious over the Reeve et al reference taken in view of Forquy et al, Lang et al and Ito et

al. Reeve et al is cited as showing carbon supported transition metal sulfide electrocatalysts such as  $Mo_xRu_yS_z$  as gas diffusion electrodes. The secondary art is applied as above.

Applicants traverse this ground of rejection as the combination of the prior art in no way teaches Applicants' invention. The Reeve et al reference relates to catalysts of MoRuS, MoRhs, MoOsS, WNus and ReRus or carbon black for oxygen reduction activities and has no relationship to CoRuS electrocatalyst for oxygen reduction. The secondary references have all been discussed with respect to their deficiencies and the combination in no way relates the problems involved and solved by Applicants' invention. Therefore, withdrawal of this rejection is requested as one cannot combine apples and oranges.

Therefore, these rejections completely fail and reversal thereof by the Board is respectfully requested.

Respectfully submitted,

  
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Charles A. Muserlian #19,683  
Attorney for Applicants  
Tel. 212 302 8989

CAM:mlp  
Enclosures

CLAIMS APPENDIXRECEIVED  
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The claims in the application are:

**Claim 1 (rejected)** An electrocatalyst for oxygen reduction comprising a cobalt and ruthenium sulfide supported on a conductive carbon black.

**Claim 2 (cancelled).**

**Claim 3 (rejected)** The electrocatalyst of claim 1 wherein said conductive carbon black is a carbon black having a surface area exceeding 120 g/m<sup>2</sup>.

**Claim 4 (cancelled).**

**Claim 5 (rejected)** The electrocatalyst of claim 1 obtained by incipient wetness impregnation of said carbon black support with an aqueous solution of precursor salts of cobalt and ruthenium, optionally comprising ruthenium chloride, drying the impregnated carbon black support and treating the resulting product under an atmosphere of hydrogen sulfide optionally diluted with an inert carrier gas.

**Claim 6 (rejected)** The electrocatalyst of claim 3 obtained by aqueous precipitation of a cobalt and ruthenium oxide on said carbon black, drying and treating

the resulting product under an atmosphere of hydrogen sulfide optionally diluted with an inert carrier gas.

**Claim 7 (rejected)** The electrocatalyst of claim 1 wherein said sulfide is a ternary sulfide of ruthenium and cobalt.

**Claims 8 and 9 (cancelled).**

**Claim 10 (rejected)** The electrocatalyst of claim 1 wherein the atomic ratio Ru:Co is comprised between 0.2 and 5.

**Claim 11 (cancelled).**

**Claim 12 (rejected)** The electrocatalyst of claim 1 obtained by incipient wetness impregnation of carbon black support with an aqueous solution of precursor salts of ruthenium and of cobalt, drying said impregnated carbon black support and treating the resulting product under an atmosphere of hydrogen sulfide optionally diluted with an inert carrier gas.

**Claim 13 (rejected)** The electrocatalyst of claim 12 wherein said precursor salts comprise at least one of  $\text{RuCl}_3$  and  $\text{Co}(\text{NO}_3)_2$ .

**Claim 14 (rejected)** The electrocatalyst of claim 12 wherein said aqueous solution of precursor salts comprises 2-propanol.

**Claim 15 (rejected)** The electrocatalyst of claim 12 wherein said drying step is carried out under vacuum at a temperature above 90°C.

**Claim 16 (rejected)** The electrocatalyst of claim 12 wherein said treatment under an atmosphere of hydrogen sulfide is carried out in a flow reactor.

**Claim 17 (rejected)** The electrocatalyst of claim 12 wherein said treatment under an atmosphere of hydrogen sulfide is carried out at a temperature exceeding 100°C.

**Claim 18 (rejected)** The electrocatalyst of claim 12 wherein said treatment under an atmosphere of hydrogen sulfide is protracted for a time exceeding thirty minutes.

**Claim 19 (rejected)** The electrocatalyst of claim 12 wherein said inert carrier gas is nitrogen and the molar ratio of said hydrogen sulfide is comprised between 0.5 and 4.

**Claim 20 (rejected)** A gas diffusion electrode comprising a conductive web wherein the catalyst of claim 1 is applied on at least one face of said conductive web.

**Claim 21 (rejected)** The gas diffusion electrode of claim 20 wherein said conductive web is a carbon cloth.

**Claim 22 (rejected)** The gas diffusion electrode of claim 20 wherein said catalyst is mixed with an optionally perfluorinated hydrophobic binder.

**Claims 23 to 35 (cancelled).**

**Claim 36 (rejected)** A method for producing a gas diffusion electrode of claim 20 comprising coating said conductive web on at least one side thereof with said catalyst optionally mixed with a first hydrophobic binder.

**Claim 37 (rejected)** The method of claim 36 wherein said conductive web is a carbon cloth.

**Claim 38 (rejected)** The method of claim 36 wherein said first hydrophobic binder is perfluorinated.

**Claim 39 (rejected)** The method of claim 36 wherein said conductive web is coated with a mixture of carbon powder and a second optionally perfluorinated hydrophobic binder on at least one side thereof, prior to said coating with said catalyst.

**Claim 40 (rejected)** The method of claim 36 further comprising a final sintering step.

**Claim 41 (allowed)** A method for producing a gas diffusion electrode comprising a conductive carbon black web wherin an electrocatalyst for oxygen reduction comprises a cobalt and ruthenium sulfide supported on carbon comprising coating a conductive web on at least one side thereof with the electrocatalyst optionally mixed with a first hydrophobic binder, subjecting the web to a sintering step by heating under a hydrogen atmosphere from room temperature up to an intermediate temperature, and subsequently heating under an inert atmosphere from said intermediate temperature up to a final temperature.

**Claim 42 (allowed)** The method of claim 41 wherein said intermediate temperature is between 100 and 120°C.

**Claim 43 (allowed)** The method of claim 41 wherein said final temperature is between 300 and 350°C.

**Claim 44 (allowed)** The method of claim 41 wherein said inert atmosphere is an argon atmosphere.

**Claim 45 (allowed)** A process of hydrochloric acid depolarized electrolysis comprising assembling the gas diffusion electrode comprising a conductive web having

an electrocatalyst comprising a cobalt and ruthenium sulfide supported on a conductive carbon black as the cathode of an electrolysis cell fed with a hydrochloric acid aqueous solution and supplying oxygen thereto while applying electrical current.

**Claim 46 (rejected)** The electrocatalyst of claim 10 wherein the atomic ratio is 2.8 to 3.2.

**Claim 47 (rejected)** The electrocatalyst of claim 17 wherein the temperature is between 300 and 500°C.

**Claim 48 (rejected)** The electrocatalyst of claim 18 wherein the time is 1 to 4 hours.

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EVIDENCE APPENDIX

Professor Faita's Declaration:

Giuseppe FAITA hereby deposes and says

I graduated with a thesis on anodes suitable for chlorine evolution at the Department of Physical Chemistry and Electrochemistry of the Università degli Studi di Milano where I am presently Associated Professor of Industrial Chemistry and Lecturer for the Courses of "Industrial Chemistry" and "Construction materials for chemical plants".

Since 1974 I began acting as technical advisor to chemical and engineering Companies in the fields of Industrial Chemistry and Electrochemistry.

I am author and co-author in 50 papers in the field of electrochemistry and Corrosion protection published in international journals, I have given 18 invited Lectures and I am a designated inventor in 17 pending or granted patents.

The above application is directed to an electrocatalyst for oxygen reduction comprising a cobalt and ruthenium sulfide supported on a conductive carbon black and a method of producing a gas diffusion electrode.

The Examiner has rejected the claims as being anticipated by the Forquy et al patent which he says discloses a catalyst comprised of ruthenium and cobalt sulfide on a support and cites active carbon as a support citing lines 22 to 38 of column 2.

Lines 27 to 30 of column 2 recites as examples of the support alumina, silica, kieselguhr, titanium, zirconium oxide, silica-alumina, thorium oxide or active carbon, none of which are electrically conductive as is known by skilled electrocatalytic engineers. As noted above, the claims require a conductive carbon black and active carbon is not conductive. This is supported by Forquy patent since the active carbon is grouped with materials which are not conductive. Active carbon does not mean conductive. Therefore, the Forquy patent does not anticipate or render obvious Applicants' catalyst as it does not teach a conductive support, much less a conductive carbon black support.

Active carbon is not carbon black.

He hereby declares that all statements made herein of own knowledge are true and that all statements made on information and belief are believed to be true; and further, that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under 18 U.S.C. § 1001 and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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Giuseppe FAITA

Date: May 14, 2007

**RELATED PROCEEDINGS APPENDIX**

None

CERTIFICATION OF FACSIMILE TRANSMISSION

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I hereby certify that this paper is being facsimile transmitted to the Patent and  
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Charles A. Muserlian  
Charles A. Muserlian #19,683

5-22-08

Serial No.: 10/830,182  
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